

Supplementary Information:

**Biobased chitosan hybrid aerogels with superior adsorption: Role of graphene
oxide in CO₂ capture**

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1. Experimental

1.1. Materials

Graphite powder (micro 850) was kindly provided by Asbury Graphite Mills, Inc. Concentrated sulfuric acid (H_2SO_4 , 98%), potassium permanganate (KMnO_4 crystal), hydrogen peroxide (H_2O_2 , 30% aqueous solution), concentrated hydrochloric acid (HCl), and glacial acetic acid (CH_3COOH) were purchased from Fisher Scientific company.

1.2. Preparation of Graphene Oxide (GO)

Graphene oxide was synthesized from natural graphite powder by the modified Hummer's method, as described in the literature [1]. In brief, 3g natural graphite powder was added to 120 ml concentrated H_2SO_4 (98%) under stirring in an ice-bath (0°C). Next, 15 g of KMnO_4 was added slowly under stirring. To control the temperature below 20°C , KMnO_4 was added gradually. The ice-bath was then taken-off and the system was heated at 35°C for 2 h. Then, 250 ml deionized water was gradually added into the system and left under stirring for another 2 h. Then a solution of 700 mL deionized water with 120 mL 30 % H_2O_2 aqueous solution was added gradually to reduce the residual KMnO_4 till no bubble appeared. The color of mixture changed into brilliant yellow along with bubbling. The mixture was filtered and washed twice with 1:10 HCl aqueous solution (1L) to remove metal ions followed by 1 L of deionized water to remove acid. Finally, the solution was filtered resulting in a yellow-brown filter cake, which was washed by deionized water until the pH value of the suspension reached near 7, and then graphene oxide was dried under vacuum for 48 h at 60°C .

1.3. Characterizations

Micro Raman scattering studies were carried out at room temperature using a Horiba Jobin-Yvon LabRam HR800 spectrometer which is equipped with a charge coupled detector and two

grating systems (600 and 1800 lines/mm). A HeNe laser ($\lambda = 632.8$ nm), with an optical power of 17 mW and a spot size of $1 \mu\text{m}^2$, was focused on the sample with an Olympus microscope. Raman shifts were calibrated with a Si (100) wafer using the 520 cm^{-1} peak.

Fourier transform infrared (FT-IR) spectra were obtained on a Bomem Michelson MB-100 FT-IR spectrometer, which was equipped with a deuterated triglycine sulfate (DTGS) detector. Thirty two scans were recorded at a resolution of 4 cm^{-1} after purging with dry air.

2. RESULTS AND DISCUSSION

2.1. Preparation of Graphene Oxide

In a typical preparation, graphene oxide is produced by oxidizing graphite (C:O ratios of approximately 2:1) [2] which ultimately disrupts the delocalized electronic structure of graphite and imparts a variety of oxygen-based chemical functionalities to the surface. The oxidation of graphite has been confirmed by Raman spectroscopy and FT-IR.

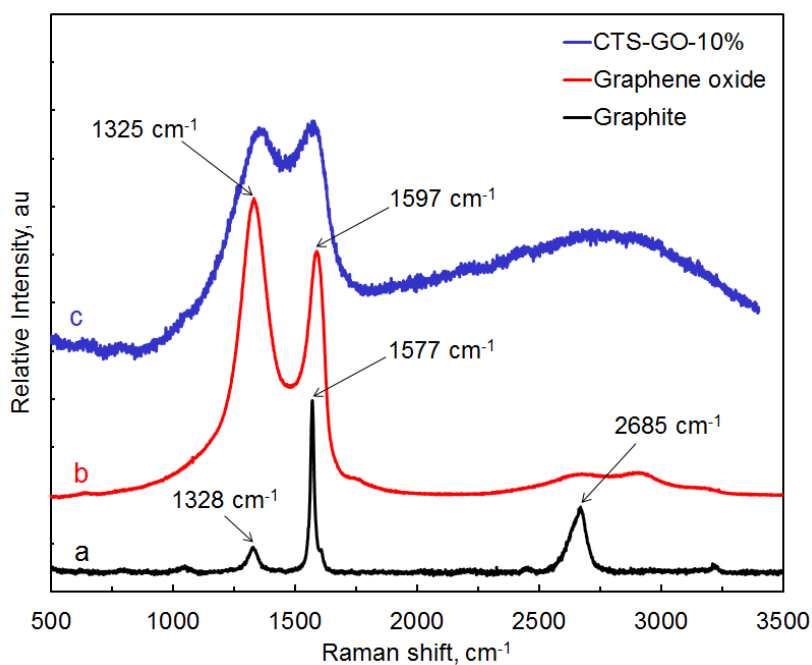


Figure S1. Raman spectra of: (a) graphite, (b) GO at 60°C , and (c) CTS-GO-10% at 400°C .

Figure S1 shows the Raman spectra of neat graphite, GO at 60 °C and CTS-GO-10% at 400 °C. As shown in Figure S1, the Raman spectrum of natural graphite exhibits strong lines at 1577 cm^{-1} and 2685 cm^{-1} , which are assigned to the G band and 2D band, respectively, while the D band is shown a weak band at 1328 cm^{-1} . The G band is attributed to the first-order scattering of the E_{2g} vibrational mode in graphite sheets and the 2D band is the overtone of the D band [3,4]. The two most intense lines appearing at 1597 and 1330 cm^{-1} for graphene oxide correspond to the G and D bands, respectively [5]. As shown in the graphene oxide spectra, The D band shifts to higher frequency and becomes prominent. The ratio of D to G band intensity (I_D/I_G) equals to 1.27, indicating the reduction in the average size of sp^2 domains due to the extensive oxidation. The 2D band of graphene oxide is seen to be broadening and decrease in relative intensity, indicating presence of defects in graphitic materials [5,6].

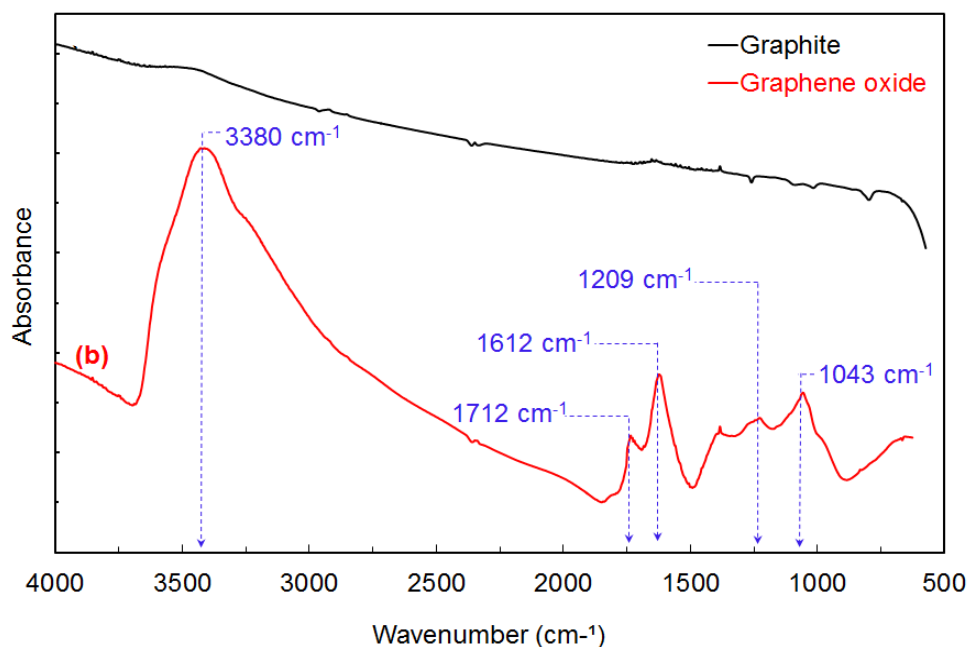


Figure S2. FT-IR spectra of: (a) graphite and (b) graphene oxide.

FT-IR results confirm the introduction of oxygen-based functional groups on the basal plane and edge of graphene. Figure S2 shows FT-IR spectra of natural graphite and neat graphene oxide. Expectedly, there is no significant peak found in the natural graphite spectrum. Otherwise, the spectrum of graphene oxide shows characteristic band at 3380 cm^{-1} (the OH stretching mode), 1720 cm^{-1} (the C=O stretching mode of carboxylic acid groups), 1612 cm^{-1} (of the skeletal vibrations of un-oxidized graphite domains), 1209 cm^{-1} (=C-OH stretching vibrations) and 1043 cm^{-1} (C-O stretching vibrations). These results are in agreement with the results reported in the literature [7-9].

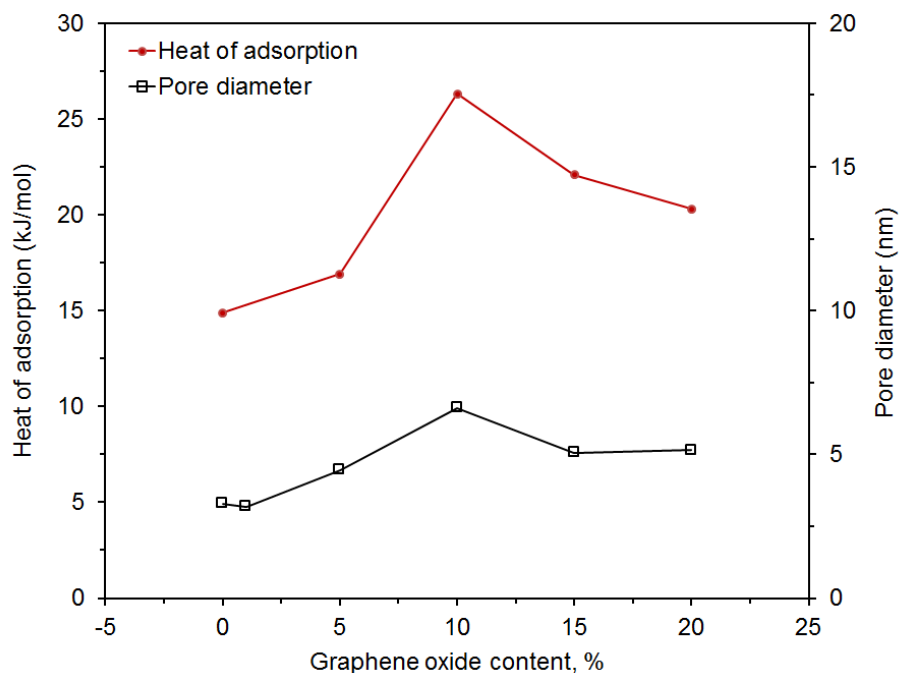


Figure S3. The relationship between the adsorption heat and the pore diameter as function of GO content.

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